

Monogamy of quantum correlations reveals frustration in quantum Ising spin system: Experimental demonstration

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We report a nuclear magnetic resonance experiment, which simulates the quantum transverse Ising spin system in a triangular lattice and further show that the monogamy of quantum correlations can be used to distinguish between the frustrated and non-frustrated phases in the ground state of this system. Adiabatic state preparation methods are used to prepare the ground states of the spin system. We employ two different multipartite quantum correlation measures to analyze the experimental ground state of the system in both the frustrated and non-frustrated regimes. In particular, we use multipartite quantum correlation measures generated by monogamy considerations of negativity, a bipartite entanglement measure, and that of quantum discord, an information-theoretic quantum correlation measure. As expected from theoretical predictions, the experimental data confirms that the non-frustrated regime shows higher quantum correlations compared to the frustrated one.

I. INTRODUCTION

The last two decades have witnessed a phenomenal growth in the domain of employing quantum correlations [1, 2] in quantum information processing tasks, with applications ranging from quantum communication [3] to measurement-based quantum computation [4] and deterministic quantum computation with a single qubit [5]. Such advantages of using quantum devices over their classical counterparts have been experimentally demonstrated in several physical systems, which include ion traps [6], photonic devices [7], nuclear magnetic resonance (NMR) [8], and cold atoms in optical lattices [9]. In particular, the Shor's factorization algorithm [10], which provides a polynomial time quantum algorithm for finding prime factors of integers, with all known classical algorithms for the same task being non-polynomial time ones, has revolutionized the developments in the fields of quantum computation and quantum cryptography. The first experimental demonstration of the Shor's algorithm was performed by NMR techniques [11].

Recently, quantum correlations have also been employed beyond its traditional ambit of quantum computation and quantum information. In particular, quantum correlation has been proposed to be a universal detector of quantum phases in quantum many-body systems [2, 12, 13]. Detecting phases of many-body systems is an important task with fundamental as well as technological implications [14]. Frustrated cooperative phenomena form one of the centerstages of the research in many-body physics. Frustration in a many-body Hamiltonian appears when it is not possible to simultaneously minimize each of the energy bonds in the Hamiltonian independently, and may appear as a result of competing interactions due to the geometry of the lattice or due to incommensurate values of the coupling strengths

[15]. Frustrated systems usually possess hugely degenerate ground states and a rich phase diagram ranging from quantum spin liquids to resonating valence-bond states [16]. Very recently, theoretical studies have shown that entanglement can be an effective tool for investigating frustrated quantum systems [17, 18]. Frustrated interactions had been known to be present in several solid state systems [16]. Recent experimental breakthroughs have made it possible to engineer frustrated spin models in ultracold atoms in optical lattices [19], trapped ions [20], NMR [21], etc, and have led to the possibility of observing the effects of entanglement in the different phases of frustrated spin models in the laboratory.

In the present work, we employ multipartite quantum correlations to distinguish the frustrated phases from the non-frustrated ones. There are many ways by which multipartite quantum correlations can be quantified [1, 2] and even though their properties can vary widely, there are some distinct connecting themes. One of them is the “monogamy” of bipartite quantum correlations, which broadly demands that if two parties are strongly quantum correlated, they cannot have a significant amount of the same with a third party [22–24].

There is a two-fold aim of the present work. First of all, we want to experimentally observe the effect of monogamy of bipartite quantum correlations in a multipartite quantum system. Secondly, we wish to apply the concept of monogamy of such correlations to distinguish between frustrated and non-frustrated phases in a lattice of quantum spins. To attain our goal, we prepare a transverse Ising spin system [25] in a triangular configuration by using NMR techniques. It undergoes a transition from a non-frustrated phase to a frustrated one when its coupling strength is varied from a ferromagnetic to an antiferromagnetic regime. We study the multipartite quantum correlations of the ground state to

detect the two different regimes. Specifically, we consider multipartite quantum correlation measures, generated from monogamy studies of bipartite quantum correlations [22–24]. In analyzing the experimentally generated ground state, we employ monogamy of (i) negativity [24, 26, 27], which is a bipartite entanglement measure, and of (ii) quantum discord [28, 29], which is an information-theoretic quantum correlation measure. For such investigations, we initially prepare the spin system in the ground state of the transverse field Hamiltonian, which is a product state and then adiabatically drive it to both frustrated and non-frustrated regimes in such a way that the system remains in the ground state of the instantaneous Hamiltonian. We then calculate the multipartite correlations of the ground state by performing quantum state tomography at different time intervals. Moreover, coinciding with the theoretical simulations, we observe that the non-frustrated regime has higher multipartite quantum correlations than the frustrated one. The transition point from the non-frustrated to the frustrated regime is well indicated by the vanishing monogamy relation.

The paper is arranged as follows. In Sec. II, we present the multipartite quantum correlations that are employed in analyzing the ground state. We describe the frustrated and non-frustrated quantum Ising spin system in Sec. III. The experimental results are presented in Sec. IV and we conclude in Sec. V.

II. MULTIPARTITE QUANTUM CORRELATIONS

Quantum correlations of a multipartite quantum state can be quantified in a variety of approaches. A prominent one among them is to use the concept of monogamy of bipartite quantum correlations [24, 26, 29]. In a tripartite scenario, monogamy of a bipartite quantum correlation restricts the amount of that correlation which can be shared between the three parties. Let us suppose that three parties, Alice, Bob, and Charu, denoted respectively as 1, 2, and 3, share a tripartite quantum state. Monogamy of quantum correlations implies that if 1 has substantial quantum correlations with 2, it can have only a restricted amount of the same with 3. To state it more precisely, for a bipartite quantum correlation measure \mathcal{Q} and a tripartite quantum state ρ_{123} , one can introduce a quantity, known as “monogamy score for \mathcal{Q} ” and denote as $\delta_{\mathcal{Q}}$, given by

$$\delta_{\mathcal{Q}}(\rho_{123}) = \mathcal{Q}_{1(23)} - \mathcal{Q}_{12} - \mathcal{Q}_{13}, \quad (1)$$

where $\mathcal{Q}_{1(23)}$ is the bipartite quantum correlation measured for the state ρ_{123} in the 1 : 23 partition. \mathcal{Q}_{12} is the same measure for the reduced state $\rho_{12} = \text{tr}_3 \rho_{123}$, and similarly for \mathcal{Q}_{13} . The three-party quantum state ρ_{123} is said to be monogamous with respect to the bipartite quantum correlation measure \mathcal{Q} , if $\delta_{\mathcal{Q}}(\rho_{123}) \geq 0$. A measure for which all tripartite quantum states produce

non-negative monogamy scores is said to be “monogamous”.

The monogamy score, and its properties, will certainly depend on the bipartite quantum correlation measure that is employed to define the score. There are many ways in which one can conceptualize bipartite quantum correlations, and correspondingly there are many bipartite quantum correlation measures [1, 2]. Broadly, these measures are defined within two paradigms, viz. the entanglement-separability paradigm and the information-theoretic one. We consider bipartite quantum correlation measures from both these paradigms to define monogamy scores, that are subsequently used for distinguishing frustrated regimes of quantum spin systems from non-frustrated ones in the experiment. As we will see below, one of these measures is monogamous for pure three-qubit quantum states, while the other is not.

Monogamy Score for Negativity Squared. – Negativity is an important quantum correlation measure for two-party quantum states [27]. It is defined within the entanglement-separability paradigm. The negativity, N_{12} , of an arbitrary bipartite quantum state ρ_{12} is the absolute value of the sum of the negative eigenvalues of the partial transposed state $\rho_{12}^{T_1}$, where the partial transposition is taken with respect to Alice (1). The importance of this measure arises from the fact that if a partially transposed bipartite quantum state has negative eigenvalues, the state must be entangled [30].

Replacing \mathcal{Q} in Eq. (1) by the squared negativity, we obtain the monogamy score for the negativity squared, given by

$$\delta_{N^2} = N_{1(23)}^2 - N_{12}^2 - N_{13}^2. \quad (2)$$

Recently, it has been shown that the negativity squared is monogamous, i.e. $\delta_{N^2} \geq 0$, for all three-qubit pure states [26]. In this paper, we measure δ_{N^2} in both the frustrated and non-frustrated regimes. For ease of reference, we call the monogamy score for negativity squared as the “entanglement monogamy score”.

Quantum Discord and Monogamy. – Let us now define another bipartite measure of quantum correlation, and importantly it does not belong to the entanglement-separability paradigm. Quantum discord is defined as the difference between two classically equivalent formulations of mutual information, when the systems involved are quantum [28], and is given by

$$D_{12} = D(\rho_{12}) = I(\rho_{12}) - J(\rho_{12}), \quad (3)$$

where $I(\rho_{12})$ and $J(\rho_{12})$ are argued to be, respectively, measures of total and classical correlations of ρ_{12} . $I(\rho_{12})$ is defined as $S(\rho_1) + S(\rho_2) - S(\rho_{12})$, where $S(\sigma) = -\text{tr}(\sigma \log_2 \sigma)$ is the von Neumann entropy of σ , $\rho_1 = \text{tr}_2 \rho_{12}$, and $\rho_2 = \text{tr}_1 \rho_{12}$. $J(\rho_{12}) = S(\rho_2) - S(\rho_{2|1})$, where the conditional entropy $S(\rho_{2|1}) = \min_{\Pi_1^1} \sum_i p_i S(\rho_{2|\Pi_1^1})$, with the minimization being performed over all possible rank-one projection-valued measurements Π_1^1 on subsystem 1. Here the output state

$\rho_{2|\Pi_i^1} = \Pi_i^1 \rho_{12} \Pi_i^1 / \text{tr}_{12}(\Pi_i^1 \rho_{12})$, and the probability $p_i = \text{Tr}(\Pi_i^1 \rho_{12})$. The monogamy score for quantum discord (referred later as the “discord monogamy score”) for a tripartite state ρ_{123} is given by [29]

$$\delta_D = D_{1(23)} - D_{12} - D_{13}. \quad (4)$$

Unlike the entanglement monogamy score, the discord monogamy score, δ_D , can be both nonnegative and negative, even for pure tripartite states [29].

III. FRUSTRATED ISING SPIN SYSTEM

Frustrated spin systems have attracted a lot of interest due to their rich phase diagrams. Moreover, current technological developments ensure the possibility of detecting such phases in the laboratory, which is also one of the goals of this paper. Frustration can be observed in the system consisting of three quantum spin-1/2 particles positioned at the corners of an equilateral triangle, and having Ising (nearest-neighbor) interactions. The Hamiltonian of this three-spin transverse Ising model is given by

$$\mathcal{H} = h(\sigma_x^1 + \sigma_x^2 + \sigma_x^3) + J(\sigma_z^1 \sigma_z^2 + \sigma_z^2 \sigma_z^3 + \sigma_z^1 \sigma_z^3), \quad (5)$$

where h is the strength of the transverse field, J is the coupling strength of the Ising interactions, and $|J| \gg h$. The three quantum spin-1/2 particles are denoted as 1, 2, 3. σ_x^i and σ_z^i , for $i = 1, 2, 3$, are the Pauli spin matrices at site i . When $J > 0$, i.e., the Ising interactions are of anti-ferromagnetic type, the system is frustrated, whereas when $J < 0$, i.e., the Ising interactions are of ferromagnetic type, the system is non-frustrated.

We now describe the adiabatic state preparation method used to prepare the ground state of this spin system in the laboratory. The quantum adiabatic theorem states that if a system is initially in the ground state and if its Hamiltonian evolves slowly with time, it will be found at any later time in the ground state of the instantaneous Hamiltonian [31]. The Hamiltonian evolution rate is governed by the relation

$$\frac{|\langle 1; t | \frac{d\mathcal{H}(t)}{dt} | 0; t \rangle|}{g^2(t)} \leq \epsilon, \quad (6)$$

where ϵ is a small number, $|0; t\rangle$ and $|1; t\rangle$ are respectively the ground and first excited states of the instantaneous Hamiltonian $\mathcal{H}(t)$, and $g(t)$ is the energy difference between the corresponding energy levels. The system stays in the instantaneous ground state of the Hamiltonian with a probability $(1 - \epsilon^2)^2$.

The spin system is initially prepared in the ground state of the Hamiltonian $h(\sigma_x^1 + \sigma_x^2 + \sigma_x^3)$. Then the system is taken to the frustrated regime by adiabatically increasing J from 0 to $|J_{max}|$ and similarly to the non-frustrated regime by increasing J from 0 to $-|J_{max}|$, where $|J_{max}| \gg h$. The system stays in the ground state

of the instantaneous Hamiltonian with high probability, if J is changed slowly enough so that it satisfies Eq. (6). The energy level diagram of the spin system is shown in the Fig. 1. The energy level of the ground state of the system is represented by red curve which is marked as E_0 . The energy level of the only excited state which is relevant in the calculation of the adiabatic evolution rate of the Hamiltonian is represented by the other red curve which is marked as E_1 . The energy levels of all the other excited states are shown in blue curves. Though there are energy levels in between E_0 and E_1 , there are no possible transitions from the ground state to these excited states, as the transition amplitudes (given by the matrix elements similar to the numerator of the left hand side in Eq. (6)) are zero in these cases.

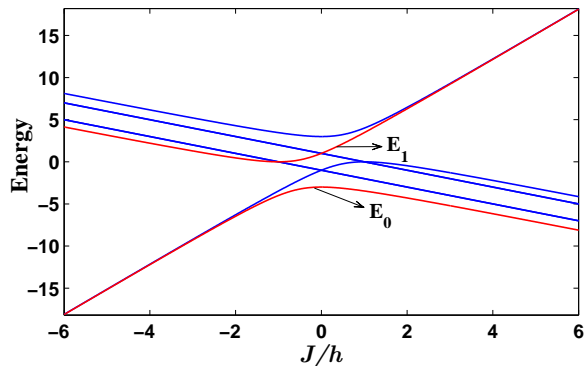


FIG. 1. Energy level diagram. The red curves, which are marked as E_0 and E_1 , represent respectively the energy levels of the ground state and the excited state which is relevant in the calculation of the adiabatic evolution rate (Eq. (6)). The blue curves represent the energy levels corresponding to the other excited states.

IV. EXPERIMENTAL IMPLEMENTATION

The spin system chosen for the experimental implementation is iodotrifluoroethylene ($\text{C}_2\text{F}_3\text{I}$) dissolved in acetone- D_6 . Here the three ^{19}F nuclear spins act as three qubits. The chemical structure of the molecule, the chemical shifts of the three fluorine nuclei, and the J -couplings between them are shown in Fig. 2. The experiments have been carried out at a temperature of 290 K in 11.7 Tesla magnetic field on a Bruker UltraShield AV III 500 MHz NMR spectrometer using a QXI probe. The ^{19}F resonance frequency at this field is 470 MHz.

In the rotating frame, the NMR Hamiltonian of the weakly coupled three-spin system is given by

$$\mathcal{H}_{\text{NMR}} = - \sum_{i=1}^3 \pi \nu_i \sigma_z^i + \sum_{i < j, i=1}^3 \frac{\pi}{2} J_{ij} \sigma_z^i \sigma_z^j. \quad (7)$$

where ν_i are the chemical shifts of the ^{19}F nuclear spins, J_{ij} are the scalar coupling constants between them. The

equilibrium density matrix under high temperature and high field approximation, is in a highly mixed state, given by

$$\rho_{eq} = \frac{1}{8}(I + \zeta \Delta\rho_{eq}), \quad (8)$$

where $\zeta \sim 10^{-5}$ is the purity factor and the deviation part of the density matrix [32]

$$\Delta\rho_{eq} \propto \gamma(\sigma_x^1 + \sigma_x^2 + \sigma_x^3). \quad (9)$$

Here γ is the gyromagnetic ratio of ^{19}F spin. In liquid state room temperature NMR, since the preparation of a pure state requires extreme conditions, it is a common practice to prepare a pseudo-pure state (PPS) that mimics the pure state [33, 34]. We have used the spatial averaging method to prepare the $|000\rangle$ PPS from the equilibrium [33].

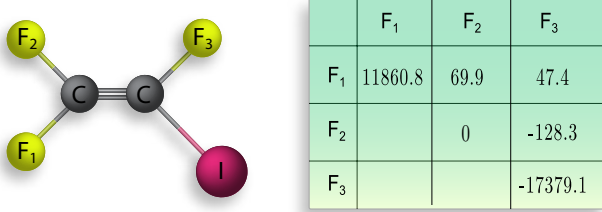


FIG. 2. Chemical structure of the molecule (left) and the table of Hamiltonian parameters (right). In the table, the diagonal elements are the chemical shifts ν_i (in Hz) of the fluorine spins and the off-diagonal elements are the scalar coupling constants J_{ij} (in Hz) between them.

In the experimental implementation, the Hamiltonian $\mathcal{H}(t)$ is discretized into $M+1$ steps as $\mathcal{H}(\frac{m}{M}T)$, where T is the total duration of the adiabatic evolution and m goes from 0 to M . The unitary operator corresponding to the each step is given by

$$U_m = \exp(-i\mathcal{H}(\frac{m}{M}T)\Delta t) = \exp(-i[h(\sigma_x^1 + \sigma_x^2 + \sigma_x^3) + J(\frac{m}{M}T)(\sigma_z^1\sigma_z^2 + \sigma_z^2\sigma_z^3 + \sigma_z^1\sigma_z^3)]\Delta t), \quad (10)$$

where $\Delta t = \frac{T}{M+1}$. The final unitary operator after the $(M+1)$ th step is given by

$$U = \prod_{m=0}^M U_m. \quad (11)$$

The unitary operator for each step can be approximated to second order in Δt by using the Trotter's formula

$$U_m = \exp(-ih(\sigma_x^1 + \sigma_x^2 + \sigma_x^3)\frac{\Delta t}{2}) \times \exp(-iJ(\frac{m}{M}T)(\sigma_z^1\sigma_z^2 + \sigma_z^2\sigma_z^3 + \sigma_z^1\sigma_z^3)\Delta t) \times \exp(-ih(\sigma_x^1 + \sigma_x^2 + \sigma_x^3)\frac{\Delta t}{2}) + \mathcal{O}(\Delta t^2). \quad (12)$$

In the experiment, the value of $h\Delta t$ was set to $\pi/21$ and that of $J(T)\Delta t$ to $\pi/4$ and $-\pi/4$ in the frustrated and

non-frustrated cases respectively. The value of $\frac{m}{M}$ was increased from 0 to 1 in 21 steps, in both the frustrated and non-frustrated cases. Considering the energy gap between the ground state (E_0) and the relevant excited state (E_1), increasing $J(t)$ linearly is not time optimal. To achieve a time-efficient adiabatic evolution, we used a sine hyperbolic variation in $J(t)$.

The ground state of the Hamiltonian at time $t = 0$, i.e. of $h(\sigma_x^1 + \sigma_x^2 + \sigma_x^3)$, is $|---\rangle$ with $|-\rangle = \frac{1}{\sqrt{2}}(|0\rangle - |1\rangle)$. This state was prepared from the $|000\rangle$ PPS by applying a $\frac{\pi}{2}$ rotation with respect to the $-y$ axis on all the three spins. This rotation was realized by a numerically optimized amplitude and phase modulated radio frequency (RF) pulse using Gradient Ascent Pulse Engineering (GRAPE) technique [36]. As evident from Eq. (11), the k^{th} step unitary operator is a product of k unitary operators (U_k s) acting on the initial state. For efficient implementation of the adiabatic evolution, we generated GRAPE pulses by cascading these unitary operators. The length of these pulses ranged between 2 ms to 30 ms. All the GRAPE pulses were optimized such that they are robust against RF field inhomogeneity and the average Hilbert-Schmidt fidelity of all these pulses are greater than 0.995. Quantum state tomography of the full density matrix was performed after every second step in both the frustrated and non-frustrated regimes. The reconstruction of the full density matrix involves an optimized set of 7 experiments and fitting of the corresponding real and imaginary parts of the single quantum spectra of all the three ^{19}F nuclear spins [37]. The real part of the reconstructed density matrices corresponding to the initial state $|---\rangle\langle---|$ and that of the final states corresponding to the last step in both the regimes are shown in the Fig. 3.

To quantitatively evaluate the experimental results, we calculate the fidelity (F) of the experimental density matrices (ρ_{exp}) with respect to the theoretical density matrices (ρ_{th}), given by

$$F = \frac{\text{tr}(\rho_{th}\rho_{exp})}{\sqrt{\text{tr}(\rho_{th}^2)\text{tr}(\rho_{exp}^2)}}. \quad (13)$$

The fidelity of the initial state $|---\rangle\langle---|$ was found to be 0.99 and that of all other final density matrices were greater than 0.984.

The entanglement monogamy score was calculated by using the relation in Eq. (2). The results are shown in Fig. 3(a). It is clear from the figure that the non-frustrated regime has higher multipartite quantum correlations as compared to the frustrated one. Except at some extreme points in the non-frustrated regime, the experimental results match with the theoretically expected curve. To qualitatively analyse the error due to the imperfect initial state, we calculated the entanglement monogamy score after applying ideal operations on the experimental initial state, and the results are also shown in Fig. 3(a). From these results, it is clear that even a small loss of fidelity of the initial state can cause large er-

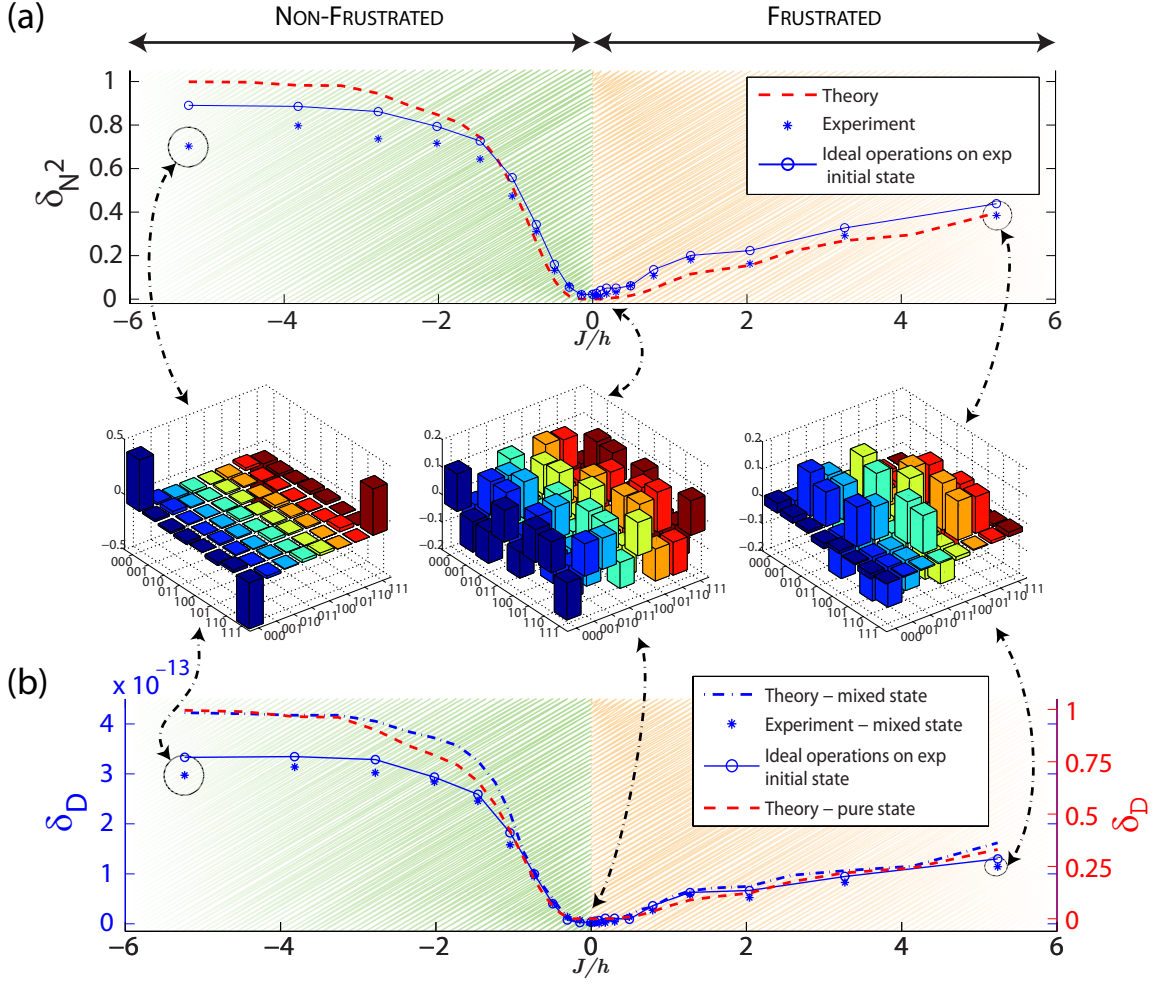


FIG. 3. (a) Entanglement monogamy scores. The red-dashed curve corresponds to the theoretically expected results, obtained by applying ideal unitary operations on the ideal initial state. The blue circles are obtained by applying the ideal unitary operations on the experimental initial state. The blue stars correspond to experimental results. (b) Discord monogamy scores. The red-dashed and blue-dash-dotted curves correspond to the theoretically expected results for the pure states and the mixed states of the NMR system respectively. These curves were obtained by applying ideal unitary operations on the ideal initial state. The blue circles correspond to the results obtained by applying ideal operations on the experimental initial state. The experimental results are shown with blue stars. The real part of the experimental density matrices are also shown for the initial state $|---\rangle\langle---|$ (middle one), the final state in the non-frustrated regime (left one), and the final state in the frustrated regime (right one).

rors in the calculation of the monogamy score. The other sources of systematic errors in the experimental data include decoherence and the non-idealities of the GRAPE pulses used to realize the unitary operators corresponding to the adiabatic evolution.

We now present our results for the monogamy of quantum discord. In this case, we make an important change in our procedure to calculate the quantum correlation of the shared states. Instead of using only the pseudo-pure part of the experimental density matrix, as was done for calculating the negativity, we used the full mixed state

density matrix of the NMR system for calculating the quantum discord. It is well known that the room temperature liquid-state NMR systems can have non-zero discord, although the purity of these systems is too small to exhibit any real entanglement [38]. The discord and the discord monogamy score were calculated using Eqs. (3) and (4) respectively. The experimental results along with the theoretically expected ones are shown in the Fig. 3(b). The theoretically expected results are shown for both pure and the NMR mixed state density matrices, where the latter's pseudo-pure part is the proportional

to the former. Although the discord monogamy scores of the mixed states are very small, their overall behavior is very much similar to that of the pure states. The results obtained by applying ideal operations on the experimental initial state are also shown in Fig. 3(b). Again we observe that the experimental data points agree with the behavior of the theoretically expected curves. From Fig. 3(b), it is clear that the non-frustrated regime again has higher multipartite quantum correlations as compared to the frustrated one. For simulating all the theoretically expected density matrices, we used the Trotter's approximation of Eq. (12) and fixed the total number of steps for the adiabatic evolution as 21 for each, in both the frustrated and non-frustrated regimes. By comparing Figs. 3(a) and 3(b), we conclude that the monogamy scores for negativity squared and quantum discord have a similar behavior in both the frustrated and non-frustrated regimes. This is despite the fact that although the two corresponding bipartite quantum correlations are defined through widely different approaches – one is via an entanglement-separability criterion and the other is through an information-theoretic paradigm. As noted before, the behavior of each one of the multipartite quantum correlation measures is different in the

frustrated and non-frustrated regimes.

V. CONCLUSION

Quantum correlation of separated quantum systems are known to be useful in a variety of phenomena. However, its detection and quantification remain difficult tasks, specially in a multipartite domain. In this work, we have been able to use quantum correlations to experimentally discern between frustrated and non-frustrated regimes of a triangular arrangement of quantum spins, in a nuclear magnetic resonance system. To attain this goal, we have used the behavior of multipartite quantum correlation measures of the ground states of this system. These multiparty measures are obtained by using the concept of monogamy of quantum correlations, which puts constraints on the sharability of quantum correlations. We believe the present study to be important not only for understanding quantum correlations in general but also in analysing various quantum phase transitions in many-body quantum systems, in particular frustrated quantum systems.

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